



⑪ Numéro de publication : **0 475 664 A1**

⑫

# **DEMANDE DE BREVET EUROPEEN**

⑳ Numéro de dépôt : **91308082.6**

⑤① Int. Cl.<sup>5</sup> : **A61K 9/02, A61K 47/10,  
A61K 47/24, A61K 47/34,  
A61K 7/48, C10M 169/04**

㉔ Date de dépôt : **04.09.91**

③① Priorité : **10.09.90 FR 9011172**

④③ Date de publication de la demande :  
**18.03.92 Bulletin 92/12**

⑥④ Etats contractants désignés :  
**DE ES FR GB IT NL SE**

⑦① Demandeur : **Dow Corning France S.A.  
Route des Cretes BP 43 Sophia Antipolis Les  
Bouillides  
F-06561 Valbonne (FR)**

⑦② Inventeur : **Aguadisch, Louis Michel Jacques  
720 A. Chemin de Villebruc  
F-06560 Valbonne (FR)  
Inventeur : Etienne, Alain  
24 Route de Pegomas  
F-06130 Grasse (FR)**

⑦④ Mandataire : **Vandamme, Luc Johan Roger  
Dow Corning Limited Cardiff Road  
Barry South Glamorgan CF6 7YL Wales (GB)**

⑤④ **Composition de lubrifiant et son utilisation.**

⑤⑦ A lubricant composition comprising (A) (i) a polydimethylsiloxane having dimethylsilanol end-blocked groups and from 3 to 51 Si atoms, and/or (ii) a silanol of the general formula  $R_2CH_2SiOH$  wherein each  $R'$  is selected from methyl and phenyl groups, at least one  $R'$  being phenyl, having dissolved therein Component (B) which is a polyoxyethylene alkylphenol of the general formula  $R-Ph-[OCH_2CH_2]_nOH$ , wherein  $R$  is selected from alkyl groups having from 6 to 12 carbon atoms,  $Ph$  represents a phenylene group and  $n$  has a value of from 8 to 10 is very useful as a clear spermicidal and virucidal lubricant composition, especially for use in condoms.

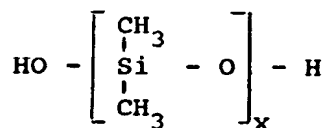
**EP 0 475 664 A1**

This invention relates to lubricant compositions and is particularly concerned with lubricant compositions having spermicidal or virucidal properties.

It is known that certain polyethyleneoxy alkylphenols have spermicidal properties and are thus useful for incorporation into lubricants for condoms. G.B. Patent Specification 1 043 513 describes such compositions wherein the polyethyleneoxy alkylphenol is dispersed in a water insoluble polysiloxane polyoxyalkylene copolymer. Although silicone liquids, in particular trimethylsiloxy end-blocked polydimethylsiloxanes, are known to lubricate elastomeric and plastic materials they are not compatible with certain compounds having spermicidal or virucidal activity. Combining silicone liquids and polyethyleneoxy alkylphenols have tended to lead to dispersions which are unstable and which have a milky, rather than a more desirable clear appearance.

We have now found that organosilicon compounds having silicon-bonded hydroxyl groups can be used as carriers for certain polyoxyethylene alkylphenols to provide spermicidal and virucidal compositions which are both stable and clear in appearance and are effective as lubricants.

According to this invention, there is provided a lubricant composition comprising an organosilicon component (A) and a polyoxyethylene alkylphenol Component (B), characterised in that Component (A) is (i) a polydimethyl siloxane of the average general formula



wherein  $x$  has a value of from 3 to 51, and/or (ii) a silanol of the general formula  $\text{R}'_2\text{CH}_3\text{SiOH}$  wherein each  $\text{R}'$  denotes a methyl or phenyl group, at least one  $\text{R}'$  being phenyl and in that Component (B) is dissolved in Component (A) and has the general formula  $\text{R-Ph-[OCH}_2\text{CH}_2\text{]}_n\text{OH}$ , wherein  $\text{R}$  represents an alkyl group having from 6 to 12 carbon atoms,  $\text{Ph}$  represents a phenylene group and  $n$  has a value of from 8 to 10.

Also included in the scope of the invention is the use of said compositions as spermicidal lubricants, especially for condoms.

Polydimethyl siloxanes which are useful as Component (A) (i) in the compositions of the present invention are substantially linear materials. They have silanol groups at the terminal positions of the polymer molecule. A particularly useful material is  $\alpha,\omega$ -dimethylsilanol end-blocked polydimethylsiloxane. Such polymers and methods for preparing them are well known in the art of silicone technology. They may

be obtained for example by the hydrolysis of dimethyldichlorosilane and, if necessary, polymerisation of the low molecular weight hydrolysis product thereof. The silanol-terminated polydimethylsiloxanes are widely employed in the silicone rubber art and differ from the so-called "silicone fluids" by the presence of terminal dimethylsilanol ( $-\text{Si}(\text{CH}_3)_2\text{OH}$ ) groups rather than trimethylsilyl ( $-\text{Si}(\text{CH}_3)_3$ ). Silanols (A) (ii) and methods for their manufacture are also well known in the silicone technology.

The preferred polydimethyl siloxanes (A) (i) are those which have from 4 to 30 silicon atoms in the chain. It is especially preferred that they have from 5 to 15 silicon atoms present in the siloxane chain. A suitable polydimethylsiloxane has the average formula  $\text{HO}[\text{Si}(\text{CH}_3)_2\text{O}]_9\text{H}$ .

The polyoxyethylene alkylphenols (B), for use in the composition according to the invention, are preferably those wherein  $n$  has a value of about 9.  $\text{R}$  may be e.g. a hexyl, heptyl or octyl group, preferably a nonyl group, for example 3,5,5-trimethylhexyl.

Lubricant compositions according to the invention are stable and clear in appearance. It is preferred that the compositions consist only of Components (A) and (B), although other components may be present. If other components are present they preferably include only small amounts, e.g. traces of polydimethylsiloxane which have terminal silanol groups, but which have a larger number of silicon atoms in the siloxane chain or of volatile silicones, e.g. polydimethyl cyclosiloxanes and hexamethyl disiloxane. It is, however, preferred for most spermicidal and virucidal applications that the polydimethylsiloxane (A) (i) is as pure as possible. This can be achieved e.g. by well known purification methods such as distillation or thin film evaporation in order to remove any volatile materials which may be present. The preferred silicone portion (A) of the composition comprises a polydimethylsiloxane (A) (i) or a mixture of two or more polydimethylsiloxanes (A) (i). Especially preferred are those compositions in which Component (A) consist only of the most preferred polydimethylsiloxanes (A) (i), i.e. those having from 5 to 15 silicon atoms.

The amount of Component (B) used in the lubricant compositions may vary from 0.1 to about 70% by weight of the combined weight of (A) and (B), more preferably from 2 to 60%. For most applications however, no more than about 50% percent by weight of (B) is used, based on the combined weight of (A) and (B). When the silanol (A) (ii) is used clear solutions result when (A) and (B) are present in any ratio; the preferred proportions of (A) (ii) and (B) are the same as those mentioned above and also apply when Component (A) (i) is used. When Component (A) (i) is used in combination with small amounts of Component (B), e.g. less than 20% by weight based on the total weight of (A) and (B), clear solutions are obtained. If higher

levels of Component (B) are to be used, such as 20% or more, best results in clarity are achieved when the preferred polydimethylsiloxanes (A) (i) are used. A more preferred range of proportions of (A) and (B) is from 2 to 25% by weight of the preferred material (B) by weight of (A) and (B) combined, most preferably from 3 to 10% by weight.

The lubricant compositions according to the invention are readily prepared by mixing Components (A) and (B) in the desired proportions. It is preferred to choose the components such that the final viscosity of the composition at 25°C would be less than 500 mm<sup>2</sup>/s, as this will ease the processibility of the composition. For most applications the preferred viscosity would be in the range of from 40 to 150 mm<sup>2</sup>/s, most preferably 50 to 100 mm<sup>2</sup>/s.

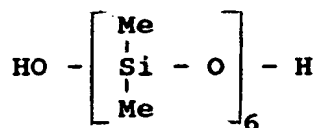
The compositions according to the invention are useful in applications requiring a liquid composition having spermicidal or virucidal properties. They are particularly suitable for use as lubricants for condoms and in other contraceptive applications. Alternative applications may include the treatment of surgical gloves or different pieces of surgical apparatus for specialised operations and treatments.

The invention accordingly also comprises condoms which use as a lubricant those compositions which are according to the invention.

The following Examples illustrate the invention. All parts and percentages are given by weight, unless otherwise indicated, and Me denotes a methyl group.

#### Example 1

8g of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 92g of a polydimethylsiloxane having the average formula



The resulting product was a clear liquid having a viscosity of 40 mm<sup>2</sup>/s at 25°C.

#### Example 2

64g of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 36g of the polydimethylsiloxane employed in Example 1. The resulting product was a clear liquid having a viscosity of 120 mm<sup>2</sup>/s at 25°C.

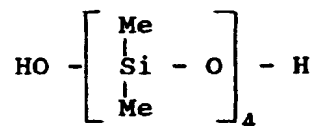
#### Example 3

10g of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 90g of

diphenylmethylsilanol. The resulting product was a clear liquid having a viscosity of 180 mm<sup>2</sup>/s at 25°C.

#### Example 4

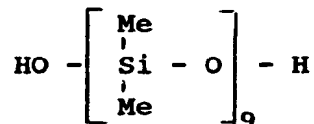
20 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 80 parts of a polydimethylsiloxane having the average formula



The resulting product was a clear liquid.

#### Example 5

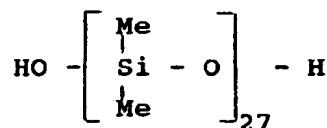
60 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 40 parts of a polydimethylsiloxane having the average formula



The resulting product was a clear liquid.

#### Example 6

4 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 96 parts of a polydimethylsiloxane having the average formula



The resulting product was a clear liquid.

#### Example 6

30 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 70 parts of a mixture of the polydimethylsiloxanes used in Examples 5 and 6 respectively, in a proportion of 8/2. The resulting product was a clear liquid having a viscosity of 60 mm<sup>2</sup>/s at 24°C.

#### Example 7

24 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 76 parts of a

mixture of the polydimethylsiloxanes used in Examples 5 and 6 respectively, in a proportion of 6/4. The resulting product was a clear liquid having a viscosity of 58 mm<sup>2</sup>/s at 24°C.

#### Example 8

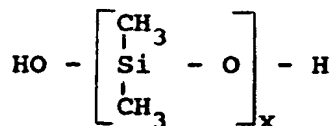
15 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 85 parts of a mixture of the polydimethylsiloxanes used in Examples 5 and 6 respectively, in a proportion of 4/6. The resulting product was a clear liquid having a viscosity of 54 mm<sup>2</sup>/s at 24°C.

#### Example 9

8 parts of polyethyleneoxy nonylphenol having 9 ethyleneoxy groups were stirred into 92 parts of a mixture of the polydimethylsiloxanes used in Examples 5 and 6 respectively, in a proportion of 8/2. The resulting product was a clear liquid having a viscosity of 51 mm<sup>2</sup>/s at 24°C.

#### Claims

1. A lubricant composition comprising an organosilicon Component (A) and a polyoxyethylene alkylphenol Component (B), characterised in that Component (A) is (i) a polydimethylsiloxane of the average general formula



wherein  $x$  has a value of from 3 to 51, and/or (ii) a silanol of the general formula  $\text{R}'_2\text{CH}_3\text{SiOH}$  wherein each  $\text{R}'$  denotes a methyl or phenyl group, at least one  $\text{R}'$  being phenyl, and in that Component (B) is dissolved in Component (A) and has the general formula  $\text{R-Ph-(OCH}_2\text{CH}_2)_n\text{OH}$ , wherein  $\text{R}$  represents an alkyl group having from 6 to 12 carbon atoms,  $\text{Ph}$  represents a phenylene group and  $n$  has a value of from 8 to 10.

2. A composition according to Claim 1 characterised in that polydimethylsiloxane (A) (i) has from 4 to 30 silicon atoms in the chain.
3. A composition according to Claim 1 or 2 characterised in that polydimethylsiloxane (A) (i) has from 5 to 15 silicon atoms present in the siloxane chain.

4. A composition according to anyone of the preceding claims characterised in that in polyoxyethylene alkylphenol (B)  $n$  has the average value of 9 and  $\text{R}$  represents a nonyl group.

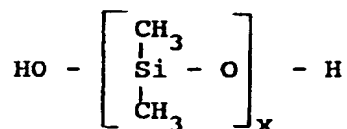
5. A composition according to anyone of the preceding claims characterised in that the percentage by weight of Component (B) based on the combined weight of (A) and (B) is from 0.1 to 70%.

6. A composition according to anyone of the preceding claims characterised in that the percentage by weight of Component (B) based on the combined weight of (A) and (B) is from 2 to 25%.

7. A composition according to anyone of the preceding claims characterised in that the final viscosity of the composition at 25°C is less than 500mm<sup>2</sup>/s.

8. A composition according to anyone of the preceding claims characterised in that the final viscosity of the composition at 25°C is in the range of from 50 to 100mm<sup>2</sup>/s.

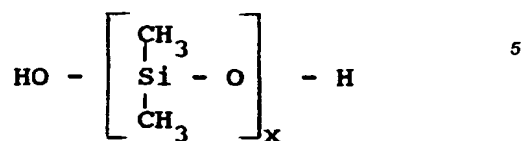
9. A method of making a lubricant compositions according to any one of the preceding claims which comprises mixing (A) (i) a polydimethyl siloxane of the average general formula



wherein  $x$  has a value of from 3 to 51, and/or (ii) a silanol of the general formula  $\text{R}'_2\text{CH}_3\text{SiOH}$  wherein each  $\text{R}'$  denotes a methyl or phenyl group, at least one  $\text{R}'$  being phenyl, with (B) a polyoxyethylene alkylphenol of the general formula  $\text{R-Ph-(OCH}_2\text{CH}_2)_n\text{OH}$  wherein  $\text{R}$  represents an alkyl group having from 6 to 12 carbon atoms,  $\text{Ph}$  represents a phenylene group and  $n$  has a value of from 8 to 10 in the desired proportions.

10. The use of a composition according to any one of Claims 1 to 9 as a spermicidal lubricant.
11. The use of a composition according to any one of Claims 1 to 9 as a spermicidal lubricant for condoms.
12. The use of a composition according to any one of Claims 1 to 9 as a virucidal lubricant.
13. A condom comprising a lubricant composition characterised in that said composition comprises

(A) (i) a polydimethylsiloxane of the average general formula



wherein  $x$  has a value of from 3 to 51, and/or (ii) 10  
 a silanol of the general formula  $\text{R}'_2\text{CH}_2\text{SiOH}$  wherein each  $\text{R}'$  denotes a methyl or phenyl group, at least one  $\text{R}'$  being phenyl, having dissolved therein (B) a polyoxyethylene alkylphenol of the general formula  $\text{R-Ph-(OCH}_2\text{CH}_2)_n\text{OH}$ , wherein  $\text{R}$  15  
 represents an alkyl group having from 6 to 12 carbon atoms,  $\text{Ph}$  represents a phenylene group and  $n$  has a value of from 8 to 10.

20

25

30

35

40

45

50

55



Office européen  
des brevets

# RAPPORT DE RECHERCHE EUROPEENNE

Numero de la demande

EP 91 30 8082

DOCUMENTS CONSIDERES COMME PERTINENTS			
Catégorie	Citation du document avec indication, en cas de besoin, des parties pertinentes	Revendication concernée	CLASSEMENT DE LA DEMANDE (Int. Cl.5)
D,A	GB-A-1 043 513 (ICI) * Revendications 1-2,6,8,10-11,21,34; page 1, formule; page 3, exemple 1 *	1,4,9- 10,13	A 61 K 9/02 A 61 K 47/10 A 61 K 47/24
A	EP-A-0 259 734 (DOW CORNING) * Revendications 1,3; page 2, formule; page 3, lignes 20-23 *	1,4,9- 10,13	A 61 K 47/34 A 61 K 7/48 C 10 M 169/04
			DOMAINES TECHNIQUES RECHERCHES (Int. Cl.5)
			A 61 K C 10 M
Le présent rapport a été établi pour toutes les revendications			
Lien de la recherche LA HAYE		Date d'achèvement de la recherche 22-10-1991	Examineur SCARPONI U.
<p>CATEGORIE DES DOCUMENTS CITES</p> <p>X : particulièrement pertinent à lui seul Y : particulièrement pertinent en combinaison avec un autre document de la même catégorie A : arrière-plan technologique O : divulgation non-écrite P : document intercalaire</p> <p>T : théorie ou principe à la base de l'invention E : document de brevet antérieur, mais publié à la date de dépôt ou après cette date D : cité dans la demande L : cité pour d'autres raisons ..... &amp; : membre de la même famille, document correspondant</p>			

EPO FORM 1503 03.82 (P0402)